

REMARKS

It is Applicants' understanding of the rejection as stated in the final Office Action, and repeated below, that the rejections of Claims 1-9 as being unpatentable over the DE 2404775 (Fuchs et al) reference, or the Konig et al reference (U.S. Patent 5,310,769) or the Keggenhoff et al reference (U.S. Patent 4,597,909) have been withdrawn. The withdrawal of these rejections is gratefully acknowledged by Applicants.

Claims 1-9 were finally rejected under 35 U.S.C. § 103(a) as being unpatentable over the GB 1203546 (Hoeschele) reference.

The Hoeschele reference, GB 1,203,546, discloses a new polyisocyanate component and a process for the production of rigid foams from this polyisocyanate composition. The polyisocyanate composition is prepared by (1) reacting at least 2.5 moles of aniline with 1 mole of formaldehyde in the presence of a mineral acid, thus forming an intermediate methylene-bridged polyphenyl polyamide mixture, (2) removing the methylenedianiline (MDA) from the intermediate polyamine mixture, thereby leaving a polyamine residue behind which contains no more than 55% by weight of methylenedianiline, and (3) phosgenating this polyamine residue to yield the corresponding polyisocyanate composition. This is described on page 1, lines 70-82. The resulting polyisocyanates are further described as inexpensive and lower in viscosity at a given MDI isocyanate than methylene-bridged polyphenyl polyisocyanate mixtures prepared by previously known methods.

The amendment filed on March 3, 2005, made it readily apparent that the presently claimed invention does **not** neutralize the acid catalyst (i.e. HCl). HCl is used in step a) of Applicants' invention to promote the reaction between the aniline and formaldehyde. One object of the present invention, was to reduce or avoid consumption of acidic catalyst, and thus, reduce or altogether eliminate the need for base to neutralize the acidic catalyst at this stage (i.e. the MDA stage) of the process. See page 4, lines 1-4 of the present application. It is further clarified in the

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present specification (see page 5, lines 16-19 and page 9, line 29 through page 10, line 3), that the process of the invention does not require neutralization of the acidic catalyst present in the polyamines of the diphenylmethane series before they are phosgenated to form the corresponding polyisocyanates. Accordingly, the presently claimed process avoids or eliminates waste water streams which contain salts and the associated reprocessing of these streams and their disposal costs.

Thus, in the presently claimed invention, the HCl that was used as a catalyst in the condensation reaction between aniline and formaldehyde is recovered during the phosgenation of the polyamines to the corresponding polyisocyanates. Then, this recovered HCl catalyst is recycled back to MDA process.

As Applicants previously pointed out, the Hoeschele reference (GB 1203546) specifically discloses at page 2, lines 56-59 that the reaction mass is neutralized after completion of the reaction between the aniline with the formaldehyde. Although Examples 1 and 2 of this reference do not expressly state that an alkaline material (e.g. sodium carbonate or sodium hydroxide) was added after completion of the aniline/formaldehyde reaction, there is no information disclosed that indicates or suggests that the hydrochloric acid used as a catalyst for this reaction was not neutralized. This is simply an assumption on the Examiner's part.

It is well known and understood by one of ordinary skill in the art that hydrochloric acid (HCl) is a by-product of the reaction between the precursor amine and phosgene which forms the desired isocyanate product. In fact, as shown in the Polyurethane Handbook, 2nd Edition, Gunter Oertel, pp.76-77 (see enclosed copy), the reaction is summarized in steps (2) and (3). In step (2), the cold phosgenation, the amine ($R-NH_2$) and phosgene ($COCl_2$) react to form the carbamoyl chloride ($R-NH-CO-Cl$) and hydrochloric acid (HCl). Then, in step (3), the hot phosgenation, the carbamoyl chloride ($R-NH-CO-Cl$) splits off hydrogen chloride (HCl) and yields the desired isocyanate ($R-NCO$). Accordingly, hydrogen chloride is a by-product of the reaction between the amine compound and phosgene to produce the desired isocyanate.

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It is also submitted that this is evident in the present specification from the discussion on page 9, lines 22-26. As set forth therein, in the process of the present invention, after completion of the reaction between the amine and phosgene, any excess phosgene, inert organic solvents and HCl are separated from the reaction mixture. It is then expressly stated that "[T]he separated HCl in this cases is made up of the HCl formed during the phosgenation of MDA with phosgene and the HCl used as catalyst for the reaction of aniline with formaldehyde to form MDA." Applicants respectfully submit that it is readily apparent from this statement that HCl is also a by-product in the present invention wherein MDA is phosgenated to form MDI.

It is therefore submitted that the working examples of the Hoeschele reference (GB 1203546) which disclose that a stream of nitrogen is swept through the reaction mixture "to remove unreacted phosgene and by-product hydrochloric acid" actually refers to the hydrochloric acid which is naturally formed in the process as a by-product. See page 3, line 108 and page 4, line 105 of the GB reference. By-products are defined, by Webster's Ninth New Collegiate Dictionary, 1990, on p. 192 (copy enclosed) as "1: something produced in a usu. industrial process in addition to the principal product 2: a secondary and sometimes unexpected or unintended result". Therefore, the term "by-product" as used in the GB reference would not be an appropriate term to describe HCl in the final product if it indeed remained in the amine mixture that was later phosgenated to form the desired isocyanate.

As Applicants have previously disclosed, and as described by the Hoeschele reference, hydrogen chloride is used as catalyst to promote the reaction between aniline and formaldehyde, thereby forming the amine mixture. See page 1, lines 72-76 of GB 1203546; and see page 5, lines 21-29 of the present specification. One of ordinary skill in the art would not refer to this hydrogen chloride as a by-product!

The Hoeschele reference clearly does not disclose or suggest the presently claimed invention to one of ordinary skill in the art. In fact, the skilled artisan has no insight into the presently claimed invention upon reading this reference. There is no information in the Hoeschele reference which would lead the skilled artisan to


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proceed in the manner Applicants have, and to eliminate the neutralization of the acidic catalyst, i.e. the hydrogen chloride. This is simply not suggested by the GB 1203546 reference.

It is respectfully submitted by Applicants that this reference does not fairly suggest the presently claimed invention to one of ordinary skill in the art. Accordingly, the rejection of Claims 1-9 under 35 U.S.C. § 103(a) in view of GB 1203546 is improper. Applicants respectfully request that this rejection be withdrawn and Claims 1-9 allowed.

Respectfully submitted,

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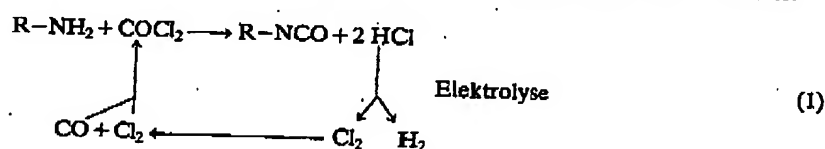
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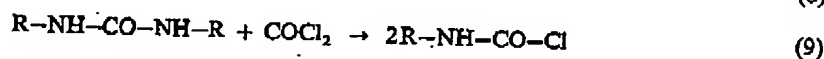
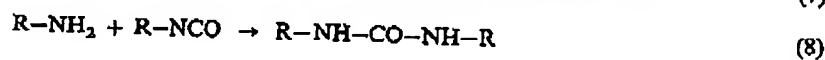
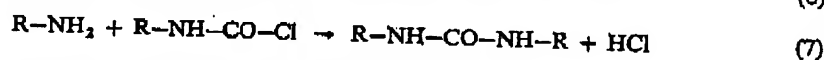
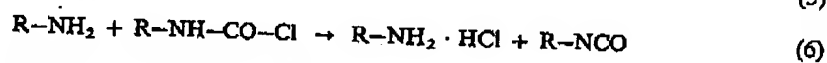
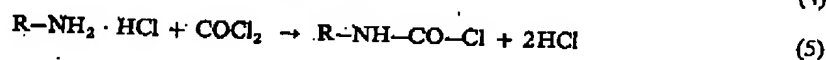
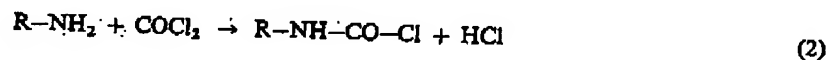
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3.2.3.1 Phosgenation

All technically important isocyanates are obtained only through the reaction of the corresponding amine with phosgene [4, 8, 11]. The phosgene needed for the reaction is continuously produced in the phosgenation unit from CO and Cl₂ and then directly reacted with the amine. Thus, the amount of phosgene present in the phosgenation unit can be reduced to a minimum. The HCl gas, which is a co-product of the reaction, is either used for other chemical reactions or reconverted to chlorine by means of HCl-electrolysis. With the complete recycling (HCl-electrolysis), the phosgenation process is self-sufficient in chlorine.



Phosgene, which was used as a war gas during World War I, has been used in large amounts for decades as an intermediate by the chemical industry. It is a colorless gas, which boils under normal pressure at 8°C. Because of its high toxicity and the correspondingly low MAK value (0,4 mg/m³), phosgenation units are equipped to meet a high technical level of safety standards. This includes, among other items, the installation of redundant, automatic, phosgene detectors, additional enclosures for phosgene containing equipment and pipes, and in case of a leak, the isolation of the total facility from the surrounding area by means of a ammonia-steam-curtain. Escaping phosgene is thus directly destroyed through the reaction with ammonia to ammonium chloride and carbonic acid. The change of reaction conditions has a substantial influence on the yield and product quality. In the complex chemical course of the process, numerous intermediate steps and by-product reactions occur. The most important:



The reaction course can be represented as follows: In the first step (cold phosgenation) the amine or the amine hydrochloride is acylated with phosgene to the carbamoyl chloride (2,5). The liberated hydrogen chloride forms with free amine the amine hydrochloride (4). This reaction is dependent on the basicity of the amine and the reaction conditions. The salt formation results in the amine group losing its basicity and thus not reacting with any NCO groups present. In the second step (hot phosgenation), the carbamoyl chloride splits off hydrochloride (3) and thus the desired isocyanate is formed. However, along with this main

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reaction the carbamoyl chloride can, especially when not enough phosgene excess is present, react with free amine to form urea and HCl (7). Urea is also formed by the reaction of an isocyanate with a free amine (8). While monofunctional amines in this case form symmetrical disubstituted ureas, the di- and polyamines give polymeric compounds. With excess phosgene the urea can be split into the carbamoyl chloride (9). As this splitting off proceeds only slowly and even at elevated temperatures is not quantitative, the urea formation must be, as much as possible, avoided [12]. For this reason excess phosgene is preferred. Further reactions of isocyanate groups such as shown in subsection 3.2.1, can lead, for example, to biurets, carbodiimides, uretdiones and isocyanates. These compounds can tie up small amounts of phosgene which in the distillation step can be split off and removed.

3.2.3.2 Phosgenation Process

The most important process for the industrial production of polyisocyanates is the phosgenation of the corresponding amine in the fluid phase at normal or slightly elevated pressure with use of a solvent [5]. The reaction with phosgene can take place both continually and discontinuously. The course of reaction is commonly separated into two temperature steps, i.e., cold and hot phosgenation. In the first step the amine is reacted with phosgene in a fast exothermic reaction at low temperatures and produces a mixture of carbamoyl chloride and amino hydrochloride. This reaction mixture is subsequently "thoroughly phosgenated" at higher temperatures until the HCl evolution is ended. In the cold phosgenation step an intensive and quick mixing of the components has a deciding influence on a smooth course of the reaction, especially for the maximization of the yield. The optimization of this process step with regard to the problems resulting from the handling of solids between 20 and 80°C was the goal of much development work. This led to the use of mixing nozzles, in-line mixers, turbulent reactors, pumps, counter current mixing chambers, and other mixing equipment with high shear effect and turbulence [4, 5]. Inert organic compounds are useful as solvents. These solvents boil below the prepared isocyanate which results in not only separation by distillation but also enables the highest possible temperature of reaction in the second phosgenation step. Chlorobenzene and ortho dichlorobenzene have been generally accepted as the most commonly used solvents. The hot phosgenation proceeds with excess phosgene at temperatures up to 180°C. It can take place in cascade of tanks, vertically or horizontally arranged pipe reactors or in packed columns, towers, etc. In this step, mixing is no longer necessary because the large amount of evolved hydrochloric acid produces sufficient turbulence. In modern continuous processes the cold phosgenation step is, as regards the apparatus, energetically distinctly separated from the hot phosgenation step. The high reaction velocity and the heat of reaction of this exothermic step can be effectively used in this way. The low pressure phosgenation in the fluid phase is illustrated in Fig. 3.2 by a simplified process flow diagram. For the sake of clarity in the illustration, the technical details and parallel installations have been eliminated. The large volume products, TDI and MDI, can be manufactured in this manner using ortho dichlorobenzene for TDI and monochlorobenzene for MDI, respectively, as solvents. The yield in the TDI process lies between 90 and 96% of the theoretical yield. With MDI inclusive its homologs the yield is 100%. Pressure processes that operate with an elevated pressure of 3 to 20 bar or higher are also known and have been demonstrated suitable for industrial scale production of isocyanates [3, 13 to 15]. They have the advantage of high reaction velocity and small reaction volumes, but the disadvantages are the known technical problems with high pressure processes. The gas-phase phosgenation is regarded as a special case. It is applicable only when the starting amine can be vaporized without decomposition [16, 17]. In this process, the reaction is accomplished by mixing the components, amine and phosgene, in the gas phase.



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backward 'bʰæ-terd-ŋ (1967): an important-sounding usu. technical word or phrase often of little meaning used chiefly to impress laymen
B, V, D, ʋ 'bʰe-ʋʱe-ʋʱe-ʋʱe *trademark* — used for underwear
B vitamin n (1940): any vitamin of the vitamin B complex
bwā-nə 'bʱwān-ə) n [Swahili, fr. Ar *abūna* our father] (1878): MASTER, BOSS
by (V) 'bi, esp before consonants **b** prep [ME, prep. & adv., fr. OE *bi*, *bē*, *bē* akin to OHG *bī*, by, near. L *ambi*- on both sides, around, Gk *amphi* (bef. 12c) 1: in proximity to: NEAR (standing ~ the window) 2: a: through or through the medium of: VIA Center ~ the door) b: in the direction of: TOWARD (north ~ east) c: into the vicinity of and beyond: PAST (went right ~ him) 3: a: during the course of (studied ~ night) b: not later than (Go ~ 2 p.m.) 4: a: through the agency or instrumentality of (forces) b: (sired) or begotten by 5: with the witness or sanction of (swear ~ all that is holy) 6: a: in conformity with (acted ~ the rules) b: ACCORDING TO (a: ways bought ~ brand) (called her ~ name) 7: with respect to 8: in or to the amount or extent of (win ~ a nose) b chiefly Scot.: in comparison with: BESIDE 9 — used as a function word to indicate successive units or increments (succeeded little ~ little) (walk two ~ two) 10 — used as a function word in multiplication, in division, and in measurements (divide a ~ b) (multiply 10 ~ 4) (a room 15 feet ~ 20 feet)
by 'bi (adv. bef. 12c) 1: close at hand: NEAR b: at or to another's home (stop ~ for a chat) 2: PAST (saw him go ~) 3: ASIDE
WAY
by 'bi, 'bi *adj* (14c) 1: being off the main route: SIDE 2: INCL. DENTIA
by or by 'bi-ŋ, n. pl *byes* 'bi:z (1567): something of secondary importance: a side issue — *by the by*, *by the way*, *incidentally*
by or by 'bi *interj* (short for goodbye) (1709) — used to express farewell; often used with following now
by-and-by 'bi-ŋ-ʱbi-ŋ (1591): a future time or occasion
by-and-by 'bi-ŋ-ʱbi-ŋ *adj* (1526): before long: soon
by and large 'bi-ŋ-ʱlɑ: (1669): on the whole; in general
by-blow 'bi-ʱbləʊ n (1594) 1: an indirect blow 2: an illegitimate child
by-blow 'bi-ŋ (alter. of *by*) (1883): the position of a participant in a tournament who has no opponent after pairs are drawn and advances to the next round without playing
byo-byo or *by-by* 'bi-ʱbi, bi-ʱbi *interj* (baby-talk redupl. of goodbye) (1736) — used to express farewell
bye-bye or *by-by* 'bi-ʱbi *adv* (1917): out esp. for a walk or ride — used with the verb go (if he wants to go ~ the baby may pat his head to indicate his desire for a hat — A. L. Gessell & Frances L. Ilg)
bye-bye or *by-by* 'bi-ʱbi n (1867): ASD. SLEEP (lie down ... and go to ~ — Rudyard Kipling)
bye-bye or *by-by* 'bi-ʱbi *adv* (1920): to bed or sleep — used with the verb go (I'll run in and read for just a second ... and then perhaps I'll go ~ — Elizabeth Cady Stanton)
by-election also *by-election* 'bi-ə-lek-ʱshən n (1880): a special election held between regular elections in order to fill a vacancy
by-gone 'bi-ʱgən also -gən *adj* (13c): gone by: PAST; esp.: OUTDATED — *bygone* n
by-law or *bye-law* 'bi-ləʊ n [ME *bylaw*, prob. fr. (assumed) ON *bylaga*, fr. ON *byr* town + *lag* law] (13c): a rule adopted by an organization chiefly for the government of its members and the regulation of its affairs
by-line 'bi-ʱlin n (1916) 1: a secondary line: SIDELINE 2: a line at the beginning of a news story, magazine article, or book giving the writer's name
by-line 'bi-ʱlin (1938): to write (an article) under a byline — *by-lined* -ʱlɪn-ə n
by-name 'bi-nəm n (16c) 1: a secondary name 2: NICKNAME
by-pass 'bi-pas n (1848) 1: a passage to one side; esp.: a deflected route usu. around a town 2: a: a channel carrying a fluid around a part and back to the main stream b: SHUNT 1b, 1c
by-pass w (1886) 1: a: to avoid by means of a bypass b: to cause to follow a bypass 2: a: to neglect or ignore usu. intentionally 1: CIRCUMVENT
by-past 'bi-past *adj* (15c): BYGONE
by-path 'bi-paθ, -pəθ n (14c): BYWAY
by-play 'bi-pli (1812): action engaged in on the side while the main action proceeds (as during a dramatic production)
by-product 'bi-prəd-ʱkt n (1857) 1: something produced in a new industrial process in addition to the principal product 2: a secondary, and sometimes unexpected or unintended result (unpleasant ~ of civilization)
byre 'bi(ə)r n [ME, fr. OE *byre*; akin to OE *bir* dwelling — *more si* *rower*] chiefly Brit. (bef. 12c): a cow barn
by-road 'bi-rəʊd n (1673): BYWAY
By-rune 'bi-rʌn-ŋ *adj* (1823): cf. relating to, or having the characteristics of the poet Byron or his writings — *By-road-easily* -li *adj* — *Byron-ism* 'bi-rən-iz-əm n
byssenosis 'bi-sə-nə-sɪs n, pl *bysses* -sɪz (NL, fr. L *byssus* of byssus, fr. Gk *byssos*: action engaged in on the side while the main action proceeds (as during a dramatic production)
byssenosis 'bi-sə-nə-sɪs n (ca. 1890): an occupational respiratory disease associated with inhalation of cotton, flax, or hemp dust and characterized initially by chest tightness, shortness of breath, dry cough and eventually by irreversible lung disease
byssus 'bi-sɪs n, pl *bysses* or *byss* 'bi-ʱsi, -ʱsi (NL, fr. ME *byssus*, fr. L fr. Gk *byssos* flax, of Sem origin; akin to Heb *byss* linen cloth) (14c) 1: a fine prob. linen cloth of ancient times 2 [NL, fr. Gk] : a tuft of long tough filaments by which some bivalve mollusks (as mussels) adhere to a surface
by-stander 'bi-staŋ-dər n (1619): one present but not taking part in a situation or event: a chance spectator
by-street 'bi-sɪrɪt n (1672): a street off a main thoroughfare: a *side street*
by 'biŋ n [perh. alter. of *white*] (ca. 1962): a group of adjacent black digits often shorter than a word that a computer processes as a unit (a 8-bit ~)
by the way *adv* (1548): in passing: INCIDENTALLY

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